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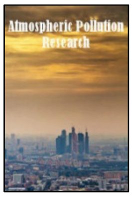

Elemental characterization of $PM_{2.5}$ and PM_1 in dense traffic area in Istanbul, Turkey

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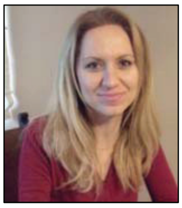
ABSTRACT

An aerosol monitoring study was conducted to measure the fine particulate matter ($PM_{2.5}$ and PM_1) concentrations and composition in the urban area of Istanbul, the most populated city in the north–west of Turkey. The sampling station was located near the D–100 highway. The PARTISOL particulate matter sampler was used during the campaign and operated from 24 April 2009 to 24 May 2009 for $PM_{2.5}$ and from 11 December 2009 to 9 April 2010 for PM_1. Nineteen $PM_{2.5}$ samples and 17 PM_1 samples were collected. The glass fiber filters were weighed before and after sampling to obtain mass concentrations. Then X–ray fluorescence was used to measure the concentration of 23 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Cu, Zn, As, Rb, Sr, Y, Mo, Ba). $PM_{2.5}$ concentration ranged between 23.8 $\mu g/m^3$ and 81.5 $\mu g/m^3$ and PM_1 concentrations were between 7.6 $\mu g/m^3$ and 30.2 $\mu g/m^3$. As a result of the principal components analysis (PCA), $PM_{2.5}$ metal emissions were dominated by significant anthropogenic sources, as expressed by high factor loadings in S, Cr, Zn, Cu and K. Crustal elements were likely related to first component (high loadings in Mg, Al, Ba and Si) for $PM_{2.5}$ and third component for PM_1.

Keywords: *$PM_{2.5}$, PM_1, XRF, heavy metals, traffic*



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1. Introduction

The chemical and physical characteristics of airborne particles are variable depending on the source of the particles. It is essentially important to know their chemical composition to evaluate and reduce the health and environmental effects of the particles. Exposure to inhalable particulate matter emissions from roadways has been implied as injurious to human health and increased risk of respiratory illnesses (Tsai et al., 2000; Lin et al., 2002). Heavy metals pollution is one of the environmental problems of particular concern to the cities which have dense residential and industrial areas and heavy traffic. The emissions of on–road motor vehicles are considered as exhaust and non–exhaust emissions. The emission of brake wear, tire wear and re–suspended road dust from roadways are non–exhaust emissions and also related to motor vehicle activity (Lough et al., 2005). Although the regulatory efforts focus on exhaust emissions, all these emissions are needed to be considered in assessing the impact of motor vehicles on human health and the environment. The observed studies about the health effects with roadway exposure may be more closely related to specific chemical components than to total mass concentrations of inhalable particles (Gavett and Koren, 2001; Claiborn et al., 2002). Metals are suspected to be linked to health impacts and therefore there is a great need to better characterize the metal emissions from motor vehicle roadways (Schaumann et al., 2004).

Most of the trace elements are determined by solution based specimen methods, such as AAS, ICP–OES and ICP–MS etc. X–ray fluorescence spectrometry (XRF) has the advantage that it can measure the solid specimen with very simple sample preparation. Recently, XRF method has been preferred to determine the chemical composition of airborne particulate matter (Carvacho et al., 2004; Lough et al., 2005; Calzolai et al., 2008; Canepari et al., 2009; Niu et al., 2010; Richard et al., 2010; Weckwerth, 2010; Apeagyei et al., 2011; Lopez et al., 2011; Indresand and Dillner, 2012). In this work, $PM_{2.5}$ and PM_1 sampling was conducted near a highway in Istanbul. $PM_{2.5}$ and PM_1 concentration levels were measured and their chemical composition was determined by a non–destructive method, XRF.

2. Material and Method

2.1. Study area

Istanbul is the most densely populated city in Turkey, located in the coastal area and separated into two parts by Istanbul Bosphorus. With an approximate population of 13 million, the metropolitan city has 11 143 industrial establishments and 2 500 000 registered motor vehicles. The study area is located in the vicinity of TEM (Trans European Motorway) and D–100 highways and the airport highway crossroad. Filter samples were collected near the D–100 highway (at the Kultur University Campus), the distance from the highway was about 25 m (Figure 1). Ataturk Airport is about 2 km away from the sampling station.

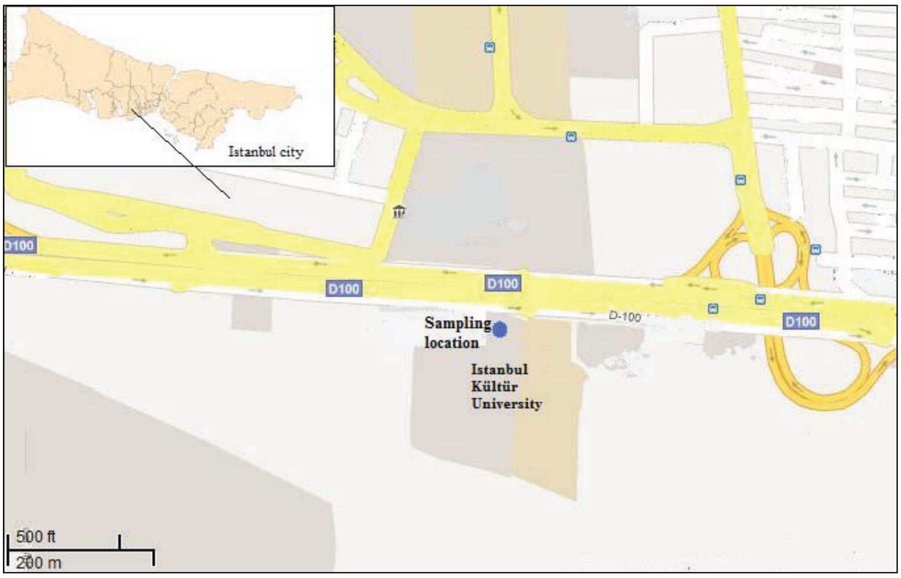

Figure 1. *Study area and the sampling location.*

2.2. Sampling procedure

$PM_{2.5}$ and PM_1 sampling was carried out using PARTISOL–FRM MODEL 2000 AIRSAMPLER (US EPA RFPS–048–117–reference method) with $PM_{2.5}$ and PM_1 inlets. Particles were collected on 47 mm glass fiber filters (Whatman, glass microfiber, GF/A) at a flow rate of 16.7 L/min. The sampling time was 24 hours. PM_1 sampling was performed randomly and 3–4 samples per month collected between 11 December 2009 and 09 April 2010. $PM_{2.5}$ sampling was done continuously during 28 April–23 May 2010. During PM_1 and $PM_{2.5}$ sampling period, the temperature was ranged between 4.1 °C and 12.4 °C; 12.0 °C and 20.0 °C, the relative humidity ranged between 67–94% and 59–91%, respectively. The filters were conditioned at a temperature of 20±1 °C and at 50±5% relative humidity for 48 h before and after field sampling and weighed on an electronic balance (RADWAG, sensitivity: 0.01 mg) to determine the mass concentrations.

2.3. XRF analysis

The quantitative analyses of Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Cu, Zn, As, Rb, Sr, Y, Mo and Ba were performed. The post weighed filter samples were analyzed using spectro IQ–2 model Energy Dispersive X–Ray Fluorescence (ED–XRF) spectrometer. The accuracy of the analytical procedures was checked against the standard reference materials. SRM 1648a (Urban particulate matter standard), IAEA–SL, IAEA–SOİL–7, BCR–176 (city waste incineration ash) and GBW 7109 standards were used. The tablets of 31 mm with 0.50 g standard and 7.95 g cellulose powders were prepared under pressure (25 tones) with dry method. The standard curves were obtained. For filter samples, 8.0 g cellulose powder tablets were prepared. Blank filter and dust filters were cut at the diameter of 31 mm and the measurements were performed. SRM 1648 standard was used for QA/QC. SRM measurement was done every 10 samples and the observed recovery was between 95% and 105%.

The SPECTRO IQ–II is equipped with an air cooled 50 W end window X–ray tube. The primary tube spectrum is monochromatized and polarized by doubly curved HOPG crystal. Two different excitation modes were chosen: Mode1 [tube voltage; 48 kV, typical power, 25 W, elements analyzed: Co–Ce (K–lines) and Hf–U (L–lines)] and Mode2 [tube voltage; 25 kV, typical power, 25 W, elements analyzed: Na–Fe (K–lines)]. A Silicon Drift Detector (SDD) was used to collect the fluorescence radiation from the sample. The resolution of the SDD was better than 175 eV for Mn Kα at an input count rate of 10 000 cps. During the measurement, the sample chamber was flushed with He. All measurement parameters are controlled by a PC connected to the system.

3. Results and Discussion

Sampling campaign was performed between 24 April 2009 and 24 May 2009 for $PM_{2.5}$ and between 11 December 2009 and 9 April 2010 for PM_1. The number of collected filters for $PM_{2.5}$ and PM_1 were 19 and 17, respectively. The mass concentrations and elemental composition of $PM_{2.5}$ and PM_1 are given in Table 1. The daily $PM_{2.5}$ concentrations varied from 23.8 μg/m^3 to 81.5 μg/m^3, and the mean $PM_{2.5}$ concentration and standard deviation was 40.5±13.7 μg/m^3. The daily PM_1 concentrations varied from 7.6 μg/m^3 to 30.2 μg/m^3, the mean PM_1 concentration and standard deviation was 22.1±6.4 μg/m^3. Most of daily concentrations (about 60% of total data) were higher than the ambient air $PM_{2.5}$ standard declared by United States Environmental Protection Agency (US EPA), given as 35 μg/m^3 for 24 h mean. PM_1 levels were higher than that measured at a background station in Finland (4.3±3.8 μg/m^3), similar to the value at the urban station of Italy (22±6 μg/m^3), but lower than at the roadside station in Hong Kong (44.0±19.4 μg/m^3) and at the urban site in China (127.3± 62.1 μg/m^3) (Ariola et al., 2006; Makkonen et al., 2010; Shen et al., 2010; Cheng et al., 2011). The value of $PM_1/PM_{2.5}$ ratio was calculated as approximately 0.55 (Table 1). The $PM_{2.5}$ and PM_1 concentrations measured in this study were two to ten times higher than those observed at the urban and background sites of Istanbul during a recent study (Sahin et al., 2012).

The emissions of particles are related to traffic as a result of fuel combustion, vehicular component wear, road degradation and roadway maintenance (Slezakova et al., 2007). The traffic related PM contains metallic elements with anthropogenic origin such as V, Cr, Fe, Ni, Cu, Zn, Pb (Sansalone and Buchberger, 1997). In some studies, Pb, Zn and Cu were indicated as marker elements of traffic emissions. The road dust include Fe, Cu, Cr, Ag, Mn, Pb, Ni, Cd, Zn (Yongming et al., 2006) and the vehicle exhaust emission include Cu, Zn, Pb, Br, Fe, Ca and Ba (Huang et al., 1994; Cadle et al., 1997). Slezakova et al. (2007) observed that the elements S, Mn, Zn, Pb, P, K and Cr originated mostly from anthropogenic activities and they were predominantly present in the fine fraction while Mg, Al, Si and Ca mostly originated from crustal sources and were predominantly present in the coarse fraction.

***Table 1**. Statistical summary of $PM_{2.5}$ and PM_1 elemental compositions (ng/m^3) at the roadside sampling station*

	$PM_{2.5}$ (N=19)		PM_1 (N=17)		
	Mean	SD	Mean	SD	$PM_1/PM_{2.5}$
Mass (µg/m^3)	40.50	13.70	22.1	6.4	0.55
Silicon (Si)	5 441	1 901	3 245	966.5	0.60
Aluminum (Al)	602.1	191.5	393.3	113.7	0.65
Calcium (Ca)	437.5	172.7	293.7	97.2	0.67
Sodium (Na)	429.5	173.8	79.8	60.9	0.19
Zinc (Zn)	384.7	197.6	108.2	80.5	0.28
Barium (Ba)	241.9	119.4	21.1	6.3	0.09
Titanium (Ti)	167.5	126.6	13.9	11.7	0.08
Crom (Cr)	121.7	40.7	5.87	3.48	0.05
Iron (Fe)	117.3	43.8	27.5	13.2	0.23
Sulfur (S)	110.5	49.4	60.0	41.4	0.54
Potassium (K)	107.4	54.3	39.0	13.7	0.36
Chlorine (Cl)	73.5	39.9	10.2	6.11	0.14
Magnesium (Mg)	66.3	36.5	31.5	10.4	0.47
Phosphorus (P)	24.0	10.2	20.2	9.31	0.84
Manganese (Mn)	42.1	51.2	1.85	1.44	0.04
Copper (Cu)	19.6	5.66	3.66	4.78	0.19
Strontium (Sr)	10.0	4.50	10.8	3.80	1.08
Vanadium (V)	2.54	2.00	0.22	0.15	0.09
Molibden (Mo)	1.08	0.44	0.09	0.10	0.09
Rubidium (Rb)	1.05	0.62	0.75	0.43	0.72
Itrium (Y)	0.81	0.38	0.13	0.12	0.16
Arsenic (As)	0.75	0.86	0.69	0.52	0.91
Cobalt (Co)	0.52	0.68	0.41	0.32	0.79

N: Number of samples, SD: Standard deviation

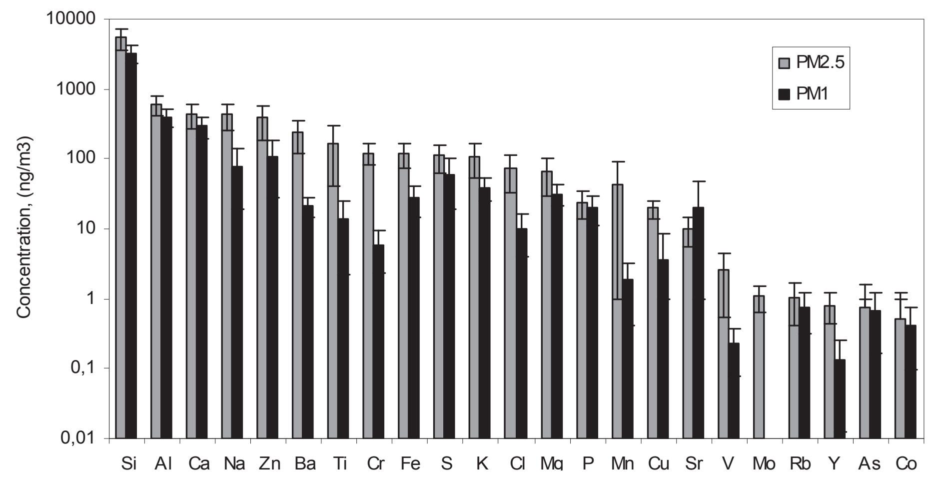

***Figure 2.** Mean elemental composition (ng/m^3) of $PM_{2.5}$ and PM_1 measured by ED-XRF.*

In this work, XRF provided the elemental composition of the measured $PM_{2.5}$ and PM_1 mass and 23 metals were determined. The comparison of metal concentration for $PM_{2.5}$ and PM_1 are illustrated in Figure 2. The contribution of the measured elements in $PM_{2.5}$ and PM_1 were ranged from 19.0% to 23.1% and from 18.1% to 20.9%, respectively. The most observed crustal element was Si, about 13.4% of $PM_{2.5}$ and 14.7% of PM_1. On average, 3.9% of $PM_{2.5}$ and 3.6% of PM_1 consisted of Al, Na, Ca and Mg. In a recent study in Istanbul (Sahin et al., 2010) it was observed that the V and Cu concentrations were between 2.0 ng/m^3–3.6 ng/m^3 and 4.6 ng/m^3–23.6 ng/m^3, respectively at a sampling station close to the traffic. These results were very similar with the results found in this study (Table 1). However, in the present study Cr and Mn concentrations of $PM_{2.5}$ were about ten times higher (62.0 ng/m^3–220.0 ng/m^3 and 13.4 ng/m^3–235.6 ng/m^3) than those in the previous study (6.4 ng/m^3–8.7 ng/m^3 and 11.8 ng/m^3–24.3 ng/m^3). In another study conducted in an urban area of Istanbul, the mean Cr concentrations in TSP (Total Suspended Particle) were observed as 279.8 ng/m^3 at a sampling station near highway and 62.2 ng/m^3 at a sampling station close to the traffic (Onat et al., 2012). In the same study, the mean V and Cu concentrations were 47.9 ng/m^3–51.9 ng/m^3 and 49.2 ng/m^3–109.7 ng/m^3, respectively. The location of the station, traffic load and airport were probably the reasons for these high levels.

Table 2. *Results of principal component analysis (PCA) in $PM_{2.5}$ and PM_1 fractions*

	$PM_{2.5}$				PM_1		
Metals	Factor 1	Factor 2	Factor 3	Factor 4	Factor 1	Factor 2	Factor 3
Na	**0.94**	-0.07	-0.09	-0.16	**0.81**	0.27	0.28
Mg	**0.85**	-0.16	0.20	0.04	**0.86**	0.04	0.48
Al	**0.87**	0.20	0.32	-0.06	0.44	0.53	**0.70**
Si	**0.92**	0.09	0.29	0.14	0.50	0.49	**0.70**
P	**0.82**	0.34	0.04	-0.11	0.04	0.53	**0.82**
S	**0.93**	-0.08	-0.05	-0.16	0.33	0.02	**0.73**
Cl	**0.65**	0.14	0.47	0.39	**0.77**	0.56	0.22
K	**0.90**	-0.06	0.25	0.12	0.60	**0.62**	0.47
Ca	-0.16	**0.82**	0.18	0,00	**0.68**	0.48	0.53
Ti	**0.90**	-0.02	-0.08	0.34	**0.74**	0.61	0.16
V	0.24	0.10	**0.68**	-0.52	**0.96**	0.22	0.13
Cr	0.87	0.27	0.11	0.16	**0.84**	0.51	0.10
Mn	0.06	-0.13	-0.04	**0.86**	**0.96**	0.18	0.10
Fe	**0.79**	0.14	0.35	-0.03	**0.88**	0.37	0.22
Co	0.39	0.36	**0.55**	0.02	0.02	**0.86**	0.26
Cu	**0.81**	0.22	**0.46**	0.11	**0.69**	0.55	0.11
Zn	**0.75**	0.02	0.15	-0.43	**0.85**	0.10	0.40
As	0.07	**0.82**	-0.17	-0.32	0.13	**0.71**	0.42
Rb	0.11	**0.87**	0.41	-0.08	0.38	**0.75**	0.47
Sr	0.87	-0.08	0.41	-0.04	-0.02	0.26	**0.91**
Y	0.33	0.87	0.07	0.22	**0.64**	0.70	0.11
Mo	**0.81**	-0.06	0.38	0.09	**0.94**	0.33	0.08
Ba	**0.56**	-0.60	0.17	0.23	**0.89**	-0.36	0.11
Total	11.5	3.8	2.3	1.8	10.7	5.6	4.7
% of Variance	50	16.4	9.8	7.9	46.4	24.3	20.2
Cumulative %	50	66.3	76.1	84	46.4	70.6	90.9
Source	Road dust and exhaust	Oil combustion	Oil combustion	Exhaust	Exhaust	Combustion	Crustal

The annual average limit value for As in respirable particles (PM_{10}) is stated as 6 ng/m^3 in EU directive (EU–Directive 99/30/EC). As and Cr are identified as carcinogenic elements and the unit risk factors are described as 0.0015 µg/m^3 for As and 1 µg/m^3 for Cr by The World Health Organization (WHO) and the Environmental Protection Agency (EPA). However, V and Mn are classified as non–carcinogenic and toxic substances with health risks. Carcinogenic substance As (0.8±0.9 ng/m^3 in $PM_{2.5}$ and 0.7±0.5 ng/m^3 in PM_1) and toxic substance Cr (19.6±5.7 ng/m^3 in $PM_{2.5}$ and 3.7±4.8 ng/m^3 in PM_1) concentrations are lower than the limit values. The average V (2.5±2.0 ng/m^3 in $PM_{2.5}$ and 0.2±0.1 ng/m^3 in PM_1) and Mn (42.2±51.1 ng/m^3 in $PM_{2.5}$ and 1.9±1.4 ng/m^3 in PM_1) concentrations are much lower than WHO guideline values of 1 and 0.15 µg/m^3.

The principal component analysis (PCA) was used for the source identification using independent variables. The PCA results are given in Table 2 and the possible pollutant sources were examined. Bold characters correspond to statistically significant variables for each factor. At the sampling site, the first component (factor 1) was extracted explaining 50% and 46.4% in $PM_{2.5}$ and PM_1 of the total variance, respectively. Factor 1 was related to anthropogenic sources, as expressed by high factor loadings in S, Cr, Zn, Cu and K. Additionally, crustal elements (Al and Si) were possibly related to first component in $PM_{2.5}$, but also related to third component in PM_1. Factor 1 is likely related to re–suspended road dust and exhaust emissions for $PM_{2.5}$, only vehicle exhaust emission for PM_1. Factor 2 explained 16.4% in $PM_{2.5}$ and 24.3% in PM_1, factor 3 39.8% in $PM_{2.5}$ and 20.2% in PM_1 and factor 4 7.9% in $PM_{2.5}$ of the total variance. Factors 2 and 3 for $PM_{2.5}$ are probably associated with oil combustion (high loading in As, medium loadings in V and Co). Also, the same was observed for factor 2 for PM_1. Factor 3 for PM_1, enriched with Al, Si, P, S and Sr, is best explained by crustal materials or road dust. Factor 4 for $PM_{2.5}$ is related to direct exhaust emissions (high loading in Mn). Also high loadings in Na, Mg, Cl (factor 1) and Ca (factor 2) can be explained by sea salt spray.

4. Conclusion

In this study, the elemental characterization of PM_1 and $PM_{2.5}$ collected at an urban station at the vicinity of the highway were determined by ED–XRF. The mean $PM_{2.5}$ and PM_1 concentrations were 40.5±13.7 µg/m^3 and 22.1±6.4 µg/m^3, respectively. These results were about ten times higher than those observed in the previous studies at the background stations in Istanbul. The fine particle exposure can be greater at the traffic microenvironments. About 20% of the $PM_{2.5}$ and PM_1 mass was constituted by 23 elements considered in this study. Carcinogenic and toxic elements concentrations (As, Cr, V and Mn) were observed at lower concentrations than the limit values of WHO and EPA. The ratios of As and Co concentrations in PM_1 and $PM_{2.5}$ as follows: As in PM_1/As in $PM_{2.5}$ was 0.91 and Co in PM_1/Co in $PM_{2.5}$ was 0.79. The PCA showed that the elements predominantly present in $PM_{2.5}$ and PM_1 originated mostly from anthropogenic activities especially traffic (exhaust and non–exhaust) emissions. The crustal elements (especially Al and Si) were dominantly present in factor 1 for $PM_{2.5}$ and in factor 3 for PM_1. In Turkey, only PM_{10} limits are defined in the national standards. Therefore, the determination of $PM_{2.5}$ and

PM_1 concentrations is very important to establish the national limits.

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